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16. ABSTRACT

As part of the development of receptor models that can be employed in air quality management, the application of target transformation factor analysis (TTFA) has been under active investigation. This method has the advantage of requiring limited a priori knowledge of the number or elemental composition of the sources contributing to the observed airborne particulate composition. The approach has been applied to aerosol composition data taken from the Regional Air Pollution Study (RAPS) of St. Louis, MO. The fine fraction at site 108 near Granite City, Illinois for July and August 1976, was examined in this study. In the data, secondary sulfate aerosol particles accounted for 48% of the mass of the average sample. Motor vehicle emissions accounted for another 29%. Flyash/soil, smelter, and an unidentified source accounted for the remaining particulate mass.

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As part of the development of receptor models that can be employed in air quality management, the application of target transformation factor analysis (TTFA) has been under active investigation. This method has the advantage of requiring limited *a priori* knowledge of the number or elemental composition of the sources contributing to the observed airborne particulate composition. The approach has been applied to aerosol composition data taken from the Regional Air Pollution Study (RAPS) of St. Louis, Mo. The fine fraction at site 108 near Granite City, Illinois for July and August, 1976, was examined in this study. In the data, secondary sulfate aerosol particles accounted for 61% of the mass of the average sample. Motor vehicle emissions accounted for another 17%. Flyash/soil, smelter, and an unidentified source accounted for the remaining particulate mass.

The application of factor analysis to source apportionment of aerosol mass[A]

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introduction

Over the decade since the promulgation of ambient air quality standards for particulate matter, there have been extensive efforts to develop strategies to meet those standards. In the initial steps, mathematical models describing the dispersion of particles from point sources were able to identify major contributors to the airborne particulate levels. However, as the increment of necessary particulate matter control became smaller, the errors inherent in these dispersion models made it unwise to rely on their continued use as the only tool for such strategy development. The use of complementary receptor models that use particulate compositional data to apportion the particle mass contributions to various sources may prove to be of great value in providing cost-efficient plans to meet air quality goals.

The initial receptor model was the chemical element balance (CEB) approach originally introduced by Miller, Friedlander, and Hidy.[1] In this model, it is assumed that the number and the elemental composition of the particle sources are known. The observed composition data are then fit to the source emission data by a multiple regression analysis where the quantity to be determined is the mass of particles attributable to each specific particle source in each sample. This approach has produced some very good fits to the data in recent studies of Washington, D.C.,[2] St. Louis, Mo.,[3] and Portland, Ore.[4] However, in order to use the CEB model, it is necessary to assume *a priori* that the number and elemental composition of the sources to be included in the analysis are known and that the emitted particle compositions do not change between the emission source and the receptor site or between the time at which the source characterization measurements were made and the ambient aerosol was sampled.

In an effort to minimize the initial assumptions, another approach to receptor models has been developed employing the multivariate statistical method, factor analysis. Factor analysis has been used to identify sources of airborne particulate matter in Boston, Mass.,[5] Tucson, Ariz.,[6] and St. Louis, Mo.[7] In these applications of factor analysis, the actual mass contribution of each of the identified sources to each sample could not be determined. Several recent studies by Hopke and co-workers[8-12] have employed a new form of factor analysis called target transformation factor analysis (TTFA). Originally developed by Malinowski and co-workers,[13] this technique permits an analysis, similar in nature to the chemical element balance method, but without the presumptions as to the number or nature of the sources in the system. Unlike the prior applications of factor analysis, TTFA allows the investigator to use his understanding of the system to determine the best representation of the actual source concentration profiles. TTFA also permits the direct calculation of the contribution of each source to each sample. This paper presents an analysis of particulate samples collected as part of the Regional Air Pollution Study (RAPS) of St. Louis, Mo. using target transformation factor analysis. By providing reliable quantitative source resolutions, without need for *a priori* assumptions, target transformation factor analysis could be an important tool in the future achievement of mandated TSP levels.

target transformation factor analysis

The fundamental assumption of factor analysis is that the amount of each element in each aerosol sample can be represented by a sum of the contributions from each of the sources that comprise the system. This model is of the form

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$$x_{ij} = \sum_{k=1}^{p} a_{ik} f_{kj} \quad (1)$$

where x_{ij} is the amount of element i in sample j and p is the number of independent sources. The contribution from each source is represented as a product of two cofactors, a_{ik} and f_{kj}. The former represents the concentration of the ith element in the particles emitted by the kth source and the latter is the amount of particulate matter per unit volume contributed by source k to sample j. The entire set of aerosol samples can be resolved into source contributions given by the matrix equation

$$X = AF \quad (2)$$

The objectives of a factor analysis are 1) To determine p, the number of independent sources that contribute to the system. 2) To identify the components of matrix A, the elemental-source profiles, and 3) To calculate F, the contribution of each source to each sample.

A matrix of sample correlations is formed from the data.

$$C = (XV)^T(XV) \quad (3)$$

where V is a diagonal matrix with the elements on the principal diagonal defined by

$$v_{ii} = [1 \sum_{j=1}^{n} x_{ij}^2]^{1/2} \quad (4)$$

This correlation matrix is then diagonalized to obtain the eigenvalues and their corresponding eigenvectors. Using several tests described in the literature,[10] the number of factors required to reproduce the data can be determined.

Target transformation rotations are used to determine the source profiles from the eigenvectors of the correlation matrix. The target transformation procedure utilizes a rotation matrix R to rotate the factor axes obtained from the diagonalization toward a set of input compositional profiles that are indicative of the emission sources for the system being studied. Rewriting equation 2 yields

$$X = ARR^{-1}F \quad (5)$$

where AR contains the representations of the concentration profiles of the real sources and $R^{-1}F$ are the mass contributions of each source to each sample. These input source profiles, called test vectors, are developed from existing knowledge of the emission profiles of various sources. Unique test vectors, having values of all but one element equal to zero are also analyzed. The uniqueness test can resolve sources that are identified primarily with only one element. To give each element equal importance in the target transformation, a weighting scheme has been added to the analysis.[14] Weighting factors for each element are the calculated variance of the element weighted with the experimental error for each datum point. The weighted rotation has greatly enhanced the ability of the target transformation to resolve sources with similar concentration profiles. It has been found that the source profiles can be developed directly from the unique vectors by an iterative process[14] so that the detailed nature of the emission profiles does not have to be initially known.

When a complete set of plausible sources has been isolated, they are scaled to reflect the actual concentration of each element in the source material. Correct scaling factors for all source profiles are determined from the calculated source contributions and the total measured sample weights by means of a multiple regression analysis.[8] The scaled test vectors are then used to calculate contributions of each source to each sample. A more detailed description of these procedures is found in Alpert and Hopke.[10]

After having established specific elemental source profiles, it is desirable to estimate the error in each parameter. Using a calculational method described by Roscoe and Hopke,[15] the matrix of differences between the original and calculated data are formed a row or column at a time

$$E = X(obs) - X(calc) \quad (6)$$

An average variance of the data in that row or column is then calculated by

TABLE I
Results of Dimensionality Tests
RAPS Station 108, Fine Particle Fraction
July and August, 1976

Factor	Eigenvalue	RMS	Chi-square	Exner	Indicator	Error
1	9.7959E+01	8.0888E+00	5.3104E+02	.266064	6.5897E−06	224.05
2	3.6110E+00	5.2483E+00	4.7528E+02	.168743	4.2825E−06	154.38
3	1.5556E+00	2.7390E+00	3.2563E+02	.101218	2.6328E−06	121.14
4	5.5105E−01	1.7396E+00	6.2843E+01	.061543	1.6412E−06	93.63
5	1.2715E−01	1.1809E+00	3.8100E+01	.047932	1.3107E−06	70.99
6	9.9126E−02	9.7390E−01	3.5213E+01	.033702	9.4530E−06	65.28
7	3.9763E−02	8.2933E−01	4.0904E+01	.025882	7.4481E−07	59.67
8	3.1119E−02	5.6344E−01	4.5305E+01	.017470	5.1594E−07	57.13
9	1.9155E−02	2.6107E−01	3.1602E+01	.008986	2.7241E−07	52.72
10	3.5141E−03	1.7963E−01	7.4681E+00	.006290	1.9580E−07	43.46
11	2.7354E−03	7.7179E−02	8.0084E+00	.002740	8.7608E−08	6.21

TABLE II
Refined Source Concentration Profiles (mg/g)
RAPS Station 108, Fine Particle Fraction, July and August, 1976

Element	Flyash/ Soil	Smelter	Motor Vehicle	Sulfate	Unknown
Al	36.3 ± 2.9	52. ± 10.	3.8 ± 2.2	0.0 ± .8	5.7 ± 3.5
Si	148. ± 8.	40. ± 23.	4.3 ± 5.4	0. ± 1.8	1.5 ± 8.4
S	.085 ± .063	195. ± 1.	4.94 ± .05	228. ± 1.	2.28 ± .08
K	15.0 ± 1.9	23. ± 6.	2.2 ± 1.5	0.0 ± .5	38.0 ± 2.3
Ca	18.4 ± 1.8	.089 ± 5.3	4.2 ± 1.3	0.0 ± .5	18.2 ± 2.1
Ti	3.89 ± .56	0.0 ± 1.8	.089 ± .422	0.0 ± .14	.15 ± .67
Mn	1.7 ± .4	2.6 ± 1.1	.32 ± .26	0.00 ± .09	1.6 ± .5
Fe	34.4 ± 1.7	54.7 ± 5.2	1.4 ± 1.3	0.0 ± .5	21.5 ± 2.0
Cu	.29 ± .50	20.2 ± 1.6	1.0 ± .4	0.00 ± .13	2.8 ± .6
Zn	.030 ± .350	203. ± 2.	.12 ± .27	0.00 ± .09	.52 ± .42
Br	1.5 ± .8	1.8 ± 2.4	17.1 ± .6	0.0 ± .19	1.3 ± .9
Pb	16. ± 5.	40. ± 15.	73.4 ± 3.6	0.0 ± 1.2	.58 ± 5.53

$$\sigma^2 = \frac{E^t W E}{m-n} \quad (7)$$

where W is the weight matrix used in the analysis, m is the number of variables and n is the number of sources used. To apportion the total variance in the data, the matrix

$$P = (A^t W A)^{-1} \quad (8)$$

is formed. The errors in the calculated results may be estimated by

$$\sigma(Y_i) = \sigma[P_{ii}]^{1/2} \quad (9)$$

where $\sigma(Y_i)$ is the estimated error in parameter Y_i. Further details may be found in the literature.[15]

description of data

The data used in this study were produced as part of the Regional Air Pollution Study (RAPS) of St. Louis, Mo. In the RAPS program, automated dichotomous samplers were operated over a two-year period at ten sites in the St. Louis metropolitan area. Over 30 000 samples were collected. Ambient aerosol samples were collected in fine, less than 2.4 μm, and coarse, 2.4 to 20 μm, fractions.[16] The samples were analyzed for total mass by beta-gauge measurements and for twenty-seven elements by x-ray fluorescence analysis.[17] This paper will present the results of a factor analysis on the fine-fraction particulate composition for samples collected during July and August, 1976, at one station. Previous studies have shown that more effective analyses can be made for samples at a single site rather than for multiple sites over a shorter time.[11] The selected station, RAPS station 108, was located near Granite City, Illinois. Filters were changed at twelve- and six-hour intervals in July and August, respectively. A total of 182 samples were produced. A majority of the determinations were below detection limits for sixty-nine samples that were eliminated from the analysis, leaving a total of 113. Of the twenty-seven elements determined for each sample, a majority of the determinations of twelve elements were below detection limits. It has been our experience that the inclusion of a large number of below detection limit points introduces sufficient noise to the system that it becomes difficult to perform the analysis and obtain meaningful results. Therefore, these twelve elements were also eliminated from the analysis. Following a closer examination of the data, only 12 elements were included in the final analysis.

results and discussion

The first step in the analysis was to determine the minimum number of factors needed to represent the data sets. The results of the tests that aid this determination are given in Table I. For each number of retained factors, the chi-square value, Exner,[19] and average percent error give an indication of the point-by-point error between the original data and the data reproduced with that number of factors. In these data, the large change in the magnitude of the eigenvalues indicated that at least four factors should be retained.

Examination of the chi-squared values shows that the maximum number of factors to be considered is six. Solutions for each number of factors are then tested to determine which gives the best fit to the data. The nature of the sources contributing to each size fraction of airborne particulate matter was determined with a target transformation analysis. The source profiles were developed by the iterative process described by Roscoe and Hopke.[14] then refined to reduce the largest single datum point errors. The target transformation analysis results for the fine fraction indicate the presence of a motor vehicle source, a sulfur source, a soil or flyash source, a smelter, and a source that cannot immediately be identified. The absolute concentrations of the test vectors used in the target transformation may not correspond exactly to the source compositions found in the St. Louis area. The predicted source profiles were scaled to the correct absolute concentrations by means of a multiple regression analysis as described by Hopke, *et al.*[8] The calculated source profiles and the associated uncertainties that

best reproduced these data are listed in Table II. The motor vehicle factor is the one with a strong dependence on Pb and Br. The calculated lead concentration in the motor vehicle factor and bromine to lead ratio of about 0.23 are typical of values reported in the literature.[19-20] The concentration of lead in motor vehicle exhaust will vary from city to city and is dependent on the ratio of leaded gasoline to unleaded and diesel powered vehicles.

The concentration of sulfur was not related to any other elements and presumably represents a secondary sulfate aerosol resulting from primary emissions of sulfur dioxide. The sulfur concentration of 22.8% in the sulfate factor is consistent with the material consisting chiefly of ammonium sulfate. The soil/flyash factor is strongly dependent on concentrations of aluminum, silicon, sulfur, potassium, calcium, and iron. The soil/flyash factor could potentially represent both soil particles as well as material originating from the combustion of coal. Differentiating soil and coal flyash, due to the similarity of their elemental profiles, is a problem often encountered in aerosol source resolution work. Coal flyash emissions are expected to contribute more to the fine fraction while crustal material should be found in the coarse fraction. Thus, it is concluded that this factor is more like that of coal flyash than soil. Reliable data for elements, such as arsenic, would be needed to clearly differentiate the contributions of soil and coal flyash.

The smelter factor is associated with the elements zinc, iron, lead, and copper. Gatz[7] noted the presence of a zinc/lead factor and attributed it to smelters located primarily in the East St. Louis and Granite City areas. There is a secondary lead smelter in Granite City, Ill. Since the factor is much higher in zinc than lead or copper, it may be that this factor is representative of the copper and zinc smelters near Sauget, Ill., farther to the south of the sampling site. Rheingrover and Gordon[21] have inferred the nature of the emissions from these sources by examination of this very large RAPS data set and by selection of the samples that are strongly impacted by single point sources. These results indicate that the zinc smelter contributes primarily to the coarse samples while the copper products plant emits fine particles that are actually higher in zinc than copper. It may then be that this factor is representative of the copper smelter.

The identity of the fifth factor is not obvious. It has high values for K, Ca, and Fe. There is a cement plant to the west of the sampling site. Small[22] has found that the concentrations of Ca and K are about the same in particles in the plume of a cement plant in Maryland. However, the values of the elemental concentrations of about 2% seem to be at least an order of magnitude too low to be cement plant particles. An alternative hypothesis is that these are particles emitted by the coking operations of the major steel production complex that is within 5 km of the sampling site. The low observed concentrations can be considered to be diluted by large quantities of carbonaceous material. There were no indications of particles high in iron that might also be expected to be emitted by that complex. Therefore, at this time a definite label cannot be assigned to this particle source.

In order to gauge the importance of each source to the amount of any particular element observed, the average airborne concentration of each element for each source has been calculated. Table III summarizes these average elemental concentrations for each source, the average predicted airborne concentration of each element, along with the average observed concentrations. The table also gives the average point-by-point percent difference between the calculated and the observed airborne concentrations. The elements, S, K, Ca, Fe, Mn, Zn, Br, and Pb, are fit very closely. The overall fit for the remaining elements is also fairly good. The average point-by-point errors in the reproduced data range up to a value of 371% for titanium. It should be noted that the average predicted and observed concentrations are very close, despite the large point-by-point error for several elements. This disparity is in part due to the tendency of the target transformation rotation to produce profiles that represent the average composition of each source. For example the average predicted and observed concentrations for copper are quite similar, but the point-by-point error indicates that this element has not been well reproduced. The

TABLE III
Summary of Mass Contributions (ng/m³)
RAPS Station 108, Fine Particle Fraction, July and August, 1976

Element	Flyash/ Soil	Smelter	Motor Vehicle	Sulfate	Unknown	Avg. Pred. Contrib.	Avg. Obs. Contrib.	Avg. % Error
Al	107	37.5	21.2	0.0	18.8	184	164	97.9
Si	436	28.7	23.8	0	5.05	493	343	132.5
S	25	140	27.6	4510	7.45	4690	4690	0
K	44.0	16.8	12.4	0.0	124	197	192	44.9
Ca	54.1	0.64	23.4	0.0	59.4	137	129	89.4
Ti	11.4	0.0	0.500	0.0	0.50	12.4	21.0	371.0
Mn	5.10	1.90	1.78	0.0	5.17	14.6	18.6	67.6
Fe	101	39.3	7.67	0.0	70.2	218	230	15.6
Cu	0.84	14.5	5.80	0.0	9.20	30.4	28.1	193.3
Zn	0.088	14.6	0.70	0.0	0.17	14.9	14.9	24.7
Br	4.54	1.30	95.3	0.0	4.10	105	98.3	16.9
Pb	47.2	28.7	410	0.0	1.89	488	538	17.8

average deviation in the mass predictions is 24%. Average deviation is the sum of the absolute values of the error in the mass prediction for each sample, divided by the number of samples. This discrepancy between the calculated and the actual masses is larger than prior studies[11] primarily because of the August samples. In August, the filters were changed every 6 hours rather than every 12 hours. The smaller mass on the filter leads to higher uncertainties in mass determinations. It may also have been the reason that fewer elements could be included in the analysis. Secondary sulfate aerosol particles account for 61% of the mass of the fine-fraction data, an average concentration of nearly 20 ng/m. Motor vehicle emissions account for another 17%. The soil/flyash averages 9.1%. The smelter contributes 2.2% and the unidentified source represents 10% of the average particulate mass.

conclusions

Target transformation factor analysis has been used to resolve sources of airborne particulate matter in samples collected as part of the RAPS study of St. Louis, Mo. Five factors were needed to represent the fine-fraction data: sulfate, motor vehicles, flyash/soil, smelter, and an unidentified source. A source profile for coal-burning power plant emissions was tentatively identified, though distinguishing crustal material from coal flyash is difficult with the available data. This distinction would be more certain if data for additional elements were available.

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